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Experimental and numerical investigations of the relation between OH* emission, flame speed and mass consumption rate

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Abstract

We investigate the relation between the intensity of the luminous emission of the excited OH* radical and the mass consumption rate of lean planar premixed methane-air flames. The flames were maintained perfectly flat using parametric acoustic stabilization in an imposed acoustic field. The consumption rate of the flames was varied by changing both the equivalence ratio and the temperature of the unburned mixture. We also compare our experimental measurements to the results of numerical simulations, using detailed chemical kinetics. For lean flames, we find that the OH* emission intensity is linear with mass consumption rate but, not proportional. Consequently, the relative fluctuation in mass consumption rate is not linearly related to the relative fluctuation in OH* emission intensity.

Introduction

Experimental investigations on combustion instabilities in general are often delicate to interpret because of the difficulty, maybe even the impossibility, of directly measuring flame burning rates, which have to be measured indirectly. Chemiluminescent emission is widely used as a diagnostic tool to characterize the local mass consumption rate and heat release rate of premixed flames. However, combustion chemiluminescence is not yet well-understood although numerous studies attempted to establish its adequacy to measure heat release rates, flame burning rates, or equivalence ratios [1-9]. The ultra-violet (UV) light emission from excited OH* radicals appears to be the most appropriate for these measurements [2,4,5,7], but there have been very few basic experimental measurements of the luminous intensity from well-controlled unstretched planar laminar flames. The purpose of this paper is to do just that.

The first paragraph explains briefly the intentions of this study, the second details the experimental apparatus and measurement techniques, and the third presents numerical tools. Finally, the results are presented and discussed in the fourth section.

Specific Objectives

The purpose of this study is to determine the relation between the intensity of chemiluminescence from excited OH* radicals and the local mass consumption rate of planar lean premixed methane-air flames. We use both direct experimental measurements on a (nearly) ideal 1-D flame, and numerical simulations.

In the experimental part, intensities of OH* luminous emission and flame speeds were measured in the same conditions, for perfectly planar unstretched flames, varying the mixture equivalence ratio and the fresh gas temperature, in order to take the principal dependences into account. We used parametric acoustic stabilization to produce planar unstretched freely propa-

gating flames [10]. This technique limits the range of equivalence ratios from 0.6 to 0.7 and a range of temperatures from 293 K to 333 K, for reasons explained below.

Numerical calculations of OH* concentrations were performed, using two different chemical kinetic schemes taken from the literature, for the same ranges of equivalence ratios and temperatures with the aim of comparing the results to our experimental measurements.

Experimental apparatus

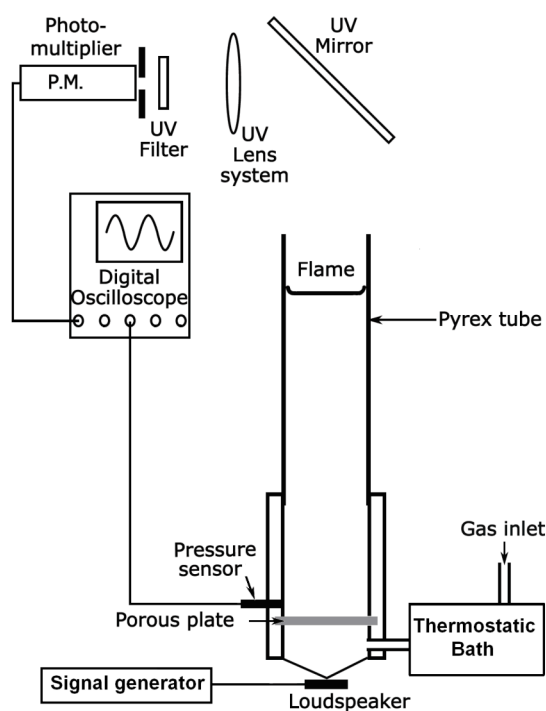


Fig. 1 - Experimental apparatus

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The experimental set-up is shown in Fig. 1. The initial flows of combustible gas (methane) and oxidant (air) were regulated using sonic nozzles. The combustible mixture was produced in excess at a constant rate. Part of the mixture was fed to the main burner and the excess mixture was consumed in a secondary burner. By this means, the flow rate in the main burner could be altered continuously without any danger of changing the composition of the mixture. The main burner consisted essentially of a half-open Pyrex tube, fed with the combustible mixture at the closed end. A porous plate made of sintered brass was placed just above the gas entry to laminarize the flow.

In order to control the temperature of the fresh gas, the mixture was circulated through a thermostatic bath just before entering the burner. The burner walls upstream from the flame were also maintained at the temperature of the fresh gas by external water circulation.

The flame was positioned not far from the tube exit and maintained perfectly stationary in the laboratory frame by carefully adjusting the gas flow rate. Using a loudspeaker, placed at the closed end of the tube, the burner was systematically excited at the lowest, quarter-wavelength, acoustic mode. The flame front was thus subjected to a periodic acoustic velocity field that served to suppress the Darrieus-Landau cellular instability and produce a perfectly flat 1-D flame [10]. This acoustic re-stabilization occurs when the amplitude of the acoustic velocity oscillation is of the order of 4 times the laminar burning velocity, and also if the burning velocity is below a critical limit. For faster flames, oscillating parametric cells appear on the flame front. For lean methane flames, the fastest flames that could be stabilized in this way had a burning speed of about 16 cm/s, which limits the ranges of equivalence ratios and fresh gas temperatures that can be explored. For this reason, our investigations were limited to mixtures with equivalence ratios in the range 0.6 to 0.7 and temperatures from 20 °C to 60 °C.

The burning velocities of the planar flames were measured by Laser Doppler Velocimetry in the presence of the stabilizing acoustic velocity field. Since the upstream flow, far from the flame front, was not a perfect plug-flow, there was a small negative velocity gradient of the order of $3\text{--}4\text{ s}^{-1}$ just ahead of the flame. The gas mean velocity was extrapolated to the mean position of the flame front. This gradient has a negligible effect on the burning velocity [11]. The burning velocities were measured independently of UV emission to eliminate any possible influence of the seeding particles on UV intensity. In general, we observed burning velocities a little lower than most values reported in the literature [12, 13, 14], but close to values published recently by Bosschaart and de Goeij [15].

The UV chemiluminescent emission from the excited radical OH*, at 307 nm, was monitored through the burned gas, as shown in Fig. 1. A UV-mirror was placed downstream from the tube exit. A pair of quartz lenses formed an image of the central region of the flame front on a diaphragm. The light then passed

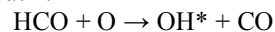
through an UV interference filter centered at 307 ± 8 nm and was detected on a photomultiplier. In order to improve the signal to noise ratio, the signal was time-averaged using a digital oscilloscope that recorded the DC level.

In all experiments, the mass consumption rate was changed by varying the fresh gas temperature and the equivalence ratio of the mixture.

Numerical investigations

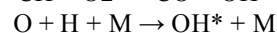
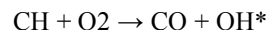
The OH* concentration of methane-air flames was calculated numerically using the PREMIX routine of the CHEMKIN package. We have used two different chemical kinetic schemes. The first scheme is that proposed by L. C. Haber [4]. His scheme is an extension of GRIMECH-3.0 and includes rates for the formation and de-excitation of the excited OH* radical. The second scheme is an extension of that proposed by A. Konnov [16]. Konnov's scheme for methane is more detailed than GRIMECH-3.0, and gives lean methane flame velocities that are in closer agreement with our experimental measurements (see Figs. 3a and 3b). We have extended Konnov's scheme by including the reactions proposed by Hall and al. [17], for production and de-excitation of the OH* radical, to which we have added the rate equation for OH* quenching with N₂, taken from the work of Haber. The thermodynamic properties of OH* were assumed to be the same as those of OH, with the adjustment for the heat of formation at 298 K taken from the work of Hall, to add the photon energy to the enthalpy of the ground state.

The originality of Haber's chemiluminescence mechanism comes from the not commonly accepted OH* formation path :



Evidence for this OH* production reaction are provided in [4]. As this reaction has never been considered, Haber has fixed the parameters for the reaction rate to match his experimental results. Also, he has modified some rates of other reactions from GRIMECH-3.0. These adjustments are of importance. For comparison, we tried to insert the OH* mechanism of Haber into the mechanism of Konnov, but the flame velocities obtained were very far from our measurements, so these results are not presented in this paper.

The OH* formation path in Hall's mechanism uses two reactions :



The first reaction is the commonly accepted production path of OH*. The second reaction is generally considered in the literature but mostly neglected. According to Hall, the alternate path for OH* formation suggested by Haber is a step not exothermic enough to produce hydroxyl radicals in an excited state. The whole OH* mechanism proposed by Hall seems to be less detailed than that proposed by Haber with 11 quenching reactions, plus one for quenching with N₂, instead of 36 in Haber's scheme. Nevertheless, it will be seen that this mechanism combined with Konnov's scheme gives

results that are in relatively good agreement with our experimental data.

In order to compare the numerical results to our measurements, all the calculations were made changing the fresh gas temperature from 293 K to 333 K, and varying the equivalence ratio of the combustible mixture from 0.6 to 0.7.

Results and discussion

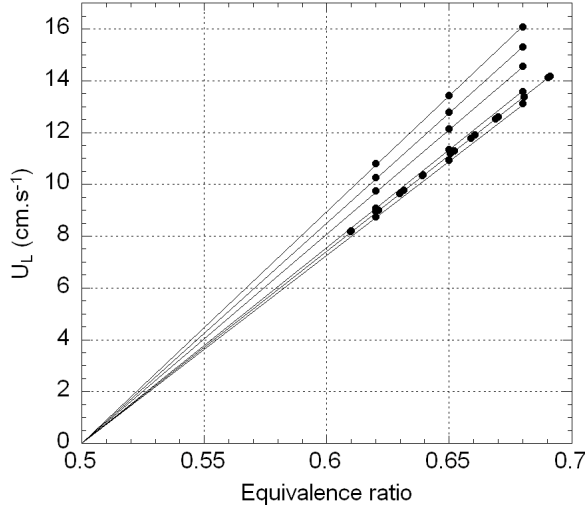


Fig. 2a - Experimental laminar burning speed as a function of the equivalence ratio for a range of fresh gas temperatures ($296 \text{ K} \leq T_u \leq 332 \text{ K}$, from bottom to top)

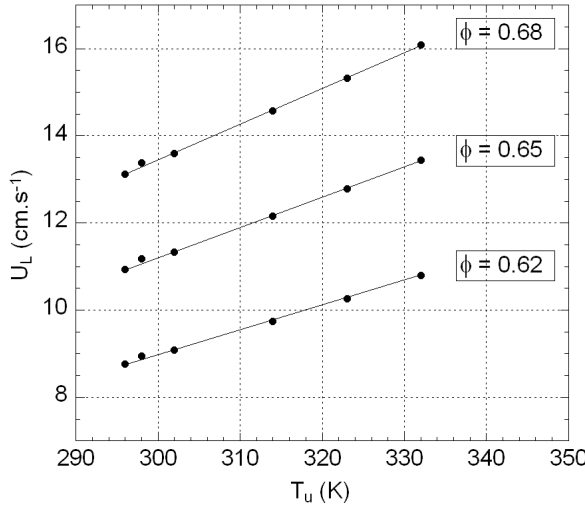


Fig. 2b - Experimental laminar burning speed as a function of the fresh gas temperature for three equivalence ratios

We first present our measurements of methane laminar flame speeds, noted U_L . The results are plotted in Figs. 2a and 2b, as functions of the equivalence ratio, ϕ , and of the fresh gas temperature, T_u , respectively. Linear relations can be observed both between the laminar flame speed and the temperature of the fresh gas, for

constant equivalence ratios, and between the laminar flame speed and the equivalence ratio of the mixture, for constant temperatures. In Fig. 2a, it is also remarkable to notice that all the extrapolations of our measurements to zero burning velocity cross the x-coordinate at equivalence ratio 0.5. We now compare these results to numerical simulations to get an indication of the relative reliability of the two kinetic schemes (see Figs. 3a and 3b).

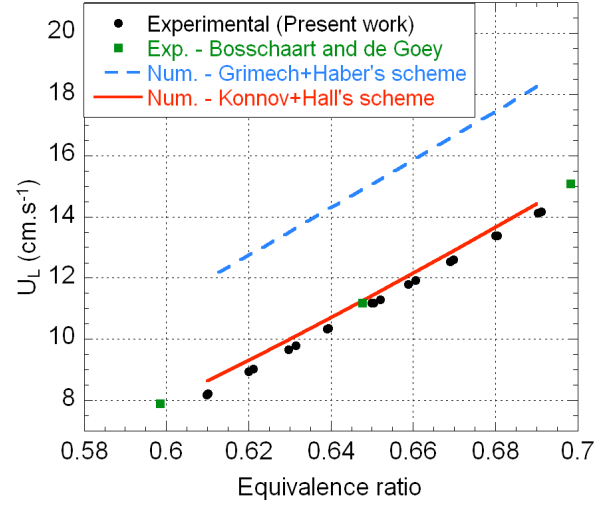


Fig. 3a - Laminar burning speed as a function of the equivalence ratio at a fresh gas temperature of 298 K

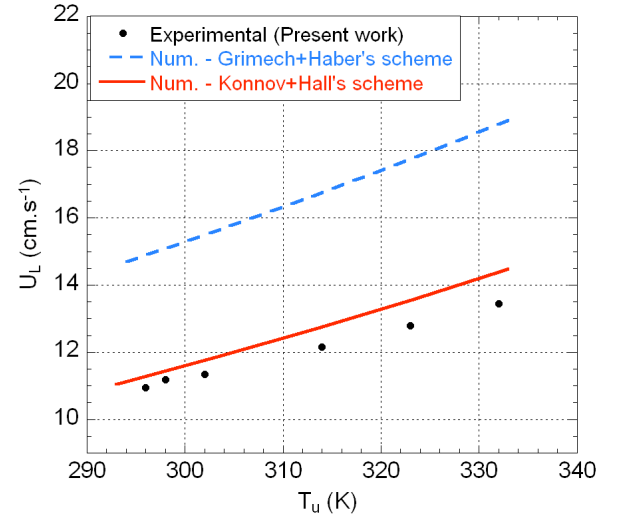


Fig. 3b - Laminar burning speed as a function of the fresh gas temperature at equivalence ratio 0.65

Fig. 3a shows the measured laminar flame speeds as a function of the equivalence ratio at the fresh gas temperature of 298 K. We compare our results both with the experimental data of Bosschaart and de Goey [15] and with the PREMIX calculations. Our results are very close to those of Bosschaart and de Goey, and to the

velocities calculated from the scheme of Konnov-Hall, whereas the laminar flame speeds given by the scheme of Grimech-Haber were found to be significantly higher. Fig. 3b shows the flame speeds as a function of the fresh mixture temperature. The velocities calculated from Konnov-Hall's scheme are still in much closer agreement with our measurements than those given by Grimech-Haber's scheme. However, the variation of the flame burning velocity with the temperature is found a little higher numerically than experimentally. These remarks are of importance since the laminar flame speed plays an essential part in the mass consumption rate of the flame, as precised later.

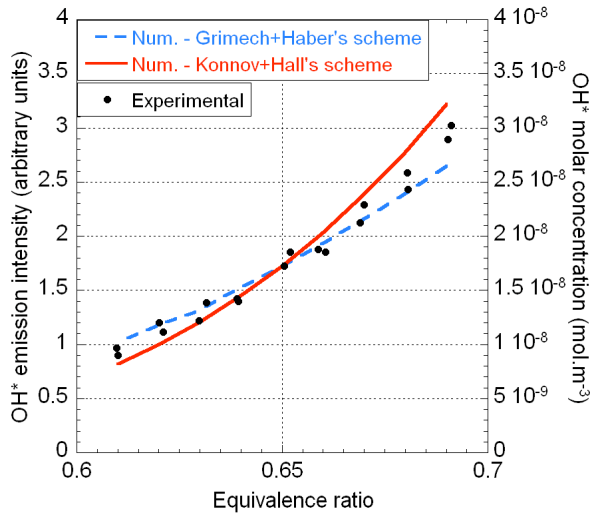


Fig. 4a – Measured OH* chemiluminescence intensity and calculated OH* molar concentration as functions of the equivalence ratio of the mixture

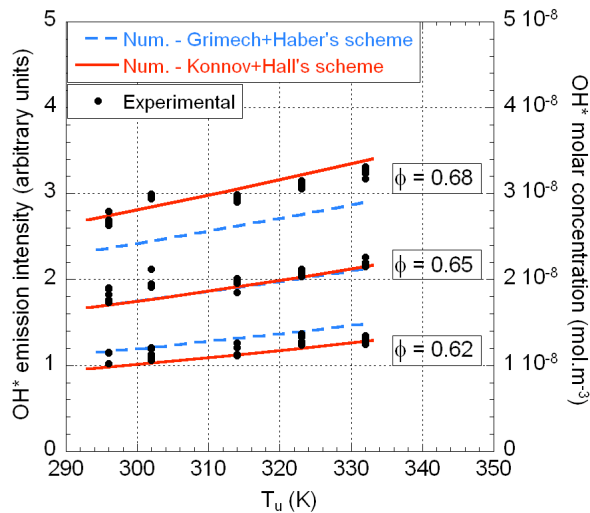


Fig. 4b - Measured OH* chemiluminescence intensity and calculated OH* molar concentration as functions of the fresh mixture temperature

We now present the raw experimental and numerical results concerning OH* chemiluminescence. Figures 4a and 4b illustrate the dependence of the measured OH* luminescence and the calculated OH* molar concentration on the equivalence ratio and on the fresh gas temperature.

In our experiments, it is not possible to calibrate the measured OH* intensity in terms of absolute OH* concentration. In order to compare the numerical and experimental results, in Figs. 4a and 4b, the scales of OH* emission intensity (left) and OH* concentration from Konnov-Hall's scheme (right) have been chosen so that they match at equivalence ratio of 0.65 and at the fresh gas temperature of 298 K. The OH* concentrations calculated with the Grimech-Haber scheme are substantially lower than those calculated with Konnov-Hall's scheme. In order to compare them with our experimental results on the same plot, we have then scaled the results from the Grimech-Haber scheme so that they also match the experimental point at the same reference equivalence ratio and temperature. To do this, the Grimech-Haber OH* concentrations have been multiplied by 2.934. The same adjustments were also applied in the following graphs. Thus, on an absolute scale, the intensities predicted using Hall's scheme, included in Konnov's, are approximately a factor 3 higher than those predicted by Haber's scheme in GRIMECH-3.0. At this stage, it is not possible to compare with absolute experimental intensities. On a relative scale, both schemes predict evolutions with equivalence ratio and with temperature that are in reasonable agreement with our experimental results.

In Fig. 4a, the relation between OH* luminescence and equivalence ratio seems to be closer to polynomial than linear, both for the experimental and numerical results. On the other hand, in Fig. 4b, the measured intensity of OH* emission appears to be linear with the temperature of the fresh gas when the equivalence ratio is held constant. This linear behavior towards temperature is also observed for the numerically calculated molar concentration of OH*, which reinforces our experimental results. Concerning the comparison of the two chemical kinetic schemes, Fig. 4a shows that Hall's scheme extended to include N₂ quenching predicts an evolution with equivalence ratio that is slightly faster than our experimental results, whereas Haber's scheme predicts a slightly slower evolution. It is difficult to say which scheme gives a better representation of our experiments. However when the fresh gas temperature is varied, Fig. 4b shows that Hall's extended scheme follows our measurements more closely than that of Haber.

In Fig. 5, we have plotted the intensity of OH* emission and the calculated OH* molar concentration as a function of the experimental mass consumption rate of the fuel, which is defined by the product $Y_f \rho U_{Lexp}$ where Y_f is the mass fraction of methane, ρ the density of the fresh mixture, and U_{Lexp} the measured laminar flame speed.

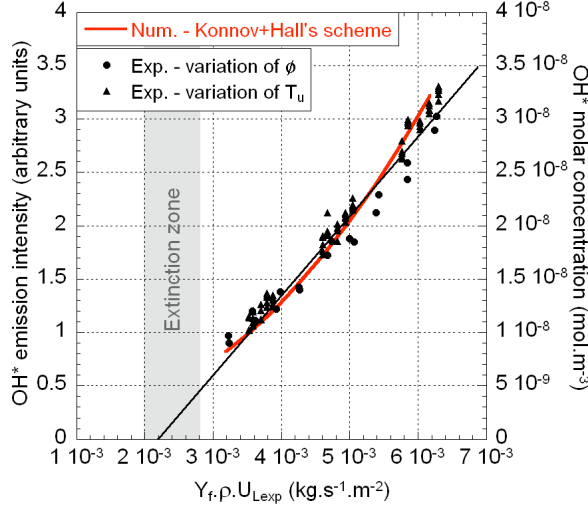


Fig. 5 - Measured OH* chemiluminescence intensity and calculated OH* molar concentration as functions of the fuel mass consumption rate

As the overall calculations with the kinetic scheme of Konnov-Hall are in better agreement with our measurements, we have chosen to not plot the numerical results found with Haber's scheme. Moreover, we have plotted only the numerical results for variation of the equivalence ratio at 298 K. The numerical results for the variation of fuel mass consumption through change of the fresh gas temperature were practically superposed, making the graph difficult to read.

Taking the experimental precision into account, it can be seen that the relation between the intensity of OH* luminous emission and the fuel mass consumption rate is almost linear and the agreement with numerical calculations is reasonable. However, it should be noted that the relation between OH* emission and fuel consumption is not proportional. The extrapolation to zero intensity of OH* chemiluminescence crosses the x-coordinate axis at a non-zero fuel consumption rate. The zero crossing is in the range of the extinction limits of methane-air flames, determined in our experimental conditions. Thus, for the lean premixed methane-air flames studied here, the relation between the chemiluminescence of the excited hydroxyl radical and the fuel mass consumption rate of the flame can be quite well approximated by the linear relation :

$$I_{OH^*} = A.(\dot{m} - \dot{m}_0), \quad (1)$$

where I_{OH^*} is the intensity of OH* luminous emission, A is a constant coefficient, \dot{m} is the fuel mass consumption rate ($Y_f \rho U_L$) and \dot{m}_0 is the fuel mass consumption rate extrapolated to zero intensity of OH* emission. This linear relation says that the variation of the fuel mass consumption rate is proportional to the variation of the intensity of OH* emission so that :

$$\delta I_{OH^*} = A. \delta \dot{m}, \quad (2)$$

and thus the relative fluctuation in mass consumption rate is related to the relative fluctuation in OH* emission through the non-linear relation :

$$\delta \dot{m} / \dot{m} = \delta I_{OH^*} / (I_{OH^*} + A.\dot{m}_0), \quad (3)$$

with \dot{m}_0 of the order of $2.2 \cdot 10^{-3} \text{ kg.s}^{-1}.\text{m}^{-2}$. It should be noted that \dot{m}_0 does not depend on the sensitivity or geometry of the luminescence detection system, but the constant A must be calibrated.

Conclusions

We studied the chemiluminescence of the hydroxyl radical in the excited state, OH*, of lean premixed methane-air flames maintained perfectly planar using the technique of parametric acoustic stabilization. The mass consumption rate has been varied, both by changing the equivalence ratio of the mixture from 0.6 to 0.7 and changing the fresh gas temperature from 293 to 333 K. In these domains, we studied the dependences of the laminar flame velocity and of the OH* luminous emission on the equivalence ratio and on the temperature of the fresh mixture. The comparison between experimental and numerical results revealed a reasonable agreement, in particular with the calculations from the kinetic mechanism proposed by Konnov extended with the scheme proposed by Hall for OH* production and quenching. Finally, we found a relation between OH* chemiluminescence and the fuel mass consumption rate of lean methane flames which can be approximated by a linear form. Nevertheless, we find that the relation is not directly proportional, and as a consequence, the relative fluctuation in mass consumption rate is not linearly related to the relative fluctuation in OH* emission intensity (see Eq. (3)).

More work is required to better understand OH* chemiluminescence and its dependences on the flame properties. We plan further experiments both with N₂-diluted methane-air flames in order to explore a larger range of equivalence ratios and also with propane-air flames. We are also planning to look more deeply into the numerical simulations, particularly concerning the kinetic mechanism of OH*.

Acknowledgements

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